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NiFe alloy particles doping effect of Gd−Ba−Cu−O bulks processed by a new cold-seeding technology

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Abstract: The process of cold seeding melt growth of $GdBa_2Cu_3O_y$ (Gd123) bulk superconductors using $NdBa_2Cu_3O_y$ (Nd123) thin films was reported. In addition, a novel cold seeding concept of combining MgO crystal and buffer pellet was also introduced. The misorientation caused by the lattice mismatch between MgO and Gd123 melt was overcome by choosing suitable heat treatment program and Gd_2BaCuO_5 (Gd211) content of the buffer pellet. The doping effect of soft ferromagnetic NiFe alloy particles was also reported. The bulk sample with 0.4% (mole fraction) doping amount shows the best performance on the flux trapping. The critical current density is largely enhanced under the external field of 1−2 T, which is promising for large-scale applications. This effect is originated from the substitution of Fe and Ni ions for the Cu sites contributing to magnetic flux pinning.

Key words: high temperature superconductor; crystal growth; cold seeding; chemical doping; magnetic flux pinning; critical current; second peak effect

1 Introduction

$REBa_2Cu_3O_y$ (RE123, RE denotes rare earth elements, e.g. Y, Gd, Sm, Nd) bulk high temperature superconductors (HTSs) possess high magnetic flux trapping performance, 17 T at 29 K as the present record [1], which is one order larger than Nd−Fe−B magnet. It makes the bulk HTSs attractive for practical applications, e.g. magnetic levitation, blushless rotating machines [2], magnetic bearings and flywheels [3]. The trapped magnetic field is proportional to the dimension and the critical current density (J_c) of the RE123 grain. Since high angle grain boundaries seriously affect the J_c, well textured single grains with large size are required.

The top-seeded melt-growth (TSMG) method has been accepted as an effective method to process RE123 single grains [4]. Considerable modifications have been achieved during the passed two decades. During the processing, the seeding procedure is a difficult point. Because of the lattice mismatch and low reactivity, single crystals, such as MgO, are not suitable to be directly used as seeds. Therefore, Nd123 or Sm123 crystals with relatively higher melting point are usually employed as seeds. Recently, MURALIDHAR et al [5] have reported the batch processing of Gd123 bulks with high performances and low costs [5]. The key point is the using of Nd123 thin films grown on MgO substrates as cold seeds to induce textured growth. Thanks to the superheating phenomenon of Nd123 thin films, a maximum temperature (T_{max}) even higher than 1090 °C could be applied, which is important for crystal growth [6]. Furthermore, it was reported that a buffer pellet, inserted between the seed and matrix, can effectively inhibit the chemical contamination caused by the dissolution of seed without affecting the texture growth of the bulk, and the T_{max} is further increased to 1096 °C [6,7].

On the other hand, refinements of Gd_2BaCuO_5 (Gd211) particles and their distribution in the matrix [8], and addition of artificial pinning centers by chemical doping are proved to be effective to enhance the J_c and irreversibility field of RE123 bulks. Various kinds of mental oxides, e.g. ZrO_2 [9,10], $RE_2Ba_4MCuO_{11}$ (M=Zr, U, Mo, W, Ta, Hf, Nb) [11] have been introduced into the RE123 matrix as the second phase particles to enhance the flux pinning. Meanwhile, new doping candidates based on different pinning mechanisms are eagerly

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expected. It is predicted that soft ferromagnetic particles, embedded in superconductors, can reduce the Lorentz force on vortexes around them, thus providing deeper potential well [12]. Recently, we have reported the enhancement of flux pinning properties by doping FeB alloy particles into Gd123 bulks [13,14].

In the present work, a novel cold-seeding method will be first introduced by combining MgO seed and a buffer pellet, and the doping effect of NiFe particles in the Gd123 bulks was reported.

2 Experimental

Commercial $GdBa_2Cu_3O_y$ (3N, Gd123) and Gd_2BaCuO_5 (3N, Gd211) powders were used as initial materials. The NiFe alloy particles with 10 μm in diameter, used for doping, were also commercial. The precursor compositions were studied as Gd123+ 40% Gd211+x% NiFe (x=0, 0.3, 0.4, 0.5 and 0.8 in mole fraction). 10% Ag_2O and 0.5% Pt were added to improve the mechanical properties and inhibit the coarsening of Gd211 particles, respectively. The pressed matrix disc of 20 mm in diameter and 10 mm in thickness was put onto ZrO_2 supporting rods inside a conventional box furnace.

Three processing methods including hot seeding using Nd123 crystal seed, cold seeding using Nd123/MgO thin film seed and cold seeding using an MgO crystal and a buffer pellet were employed to grow Gd123 bulks. For hot seeding, the pre-forms were first heated to 1090 °C (T_{max}) within 10 h and held for 1 h. Then the temperature was decreased to 1020 °C for seeding. A small Nd123 crystal was put on the surface of the pre-form at this moment. After that the temperature was decreased to 1010 °C in 30 min, from where the textured growth started (T_s), and further slowly decreased by 40 °C with a cooling rate of 0.3 °C/h. In this case, the peritectic decomposition temperature (T_p) of the matrix was chosen as T_s. Finally, the temperature was decreased to room temperature in 10 h.

For cold seeding, a 2 mm×2 mm NdBCO thin film, purchased from THEVA Company, was placed on the top of the pre-form at the beginning. The heat treatment profile was performed the same as hot seeding, except removing the seeding procedure.

For the novel cold-seeding method, a small buffer pellet pressed from Gd123 powders with suitable amount of Gd211 addition was inserted between the matrix and MgO single-crystal seed (Fig. 1). The growth processing was divided into two stages: First to texture the buffer pellet and then to induce the textured growth of the matrix from the buffer pellet. This meant that after holding the pre-form at T_{max} for 1 h, the temperature was first decreased to $T_{s\text{-pellet}}$ to grow the buffer pellet and then decreased to $T_{s\text{-matrix}}$ to perform the textured growth

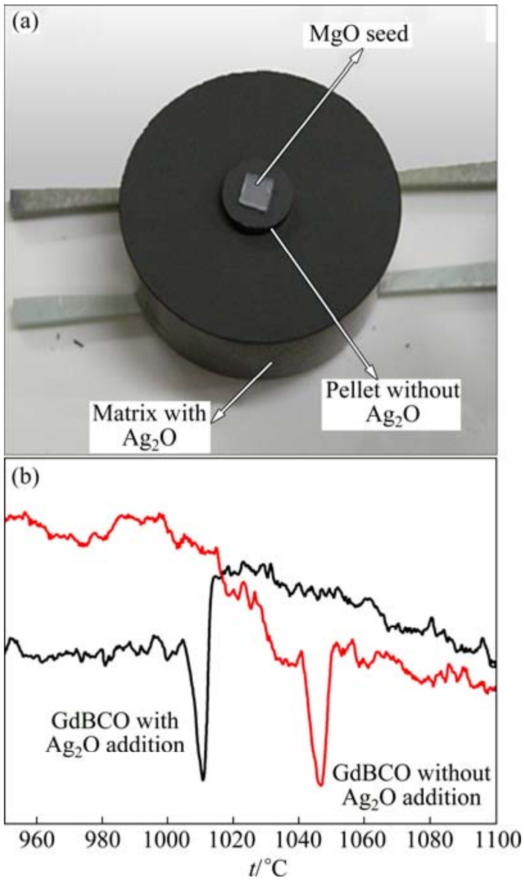

Fig. 1 Schematic illustration of arrangement of novel cold seeding method (a) and DTA traces of buffer pellet (b)

of the bulk.

Small specimens with the dimensions of 2 mm× 2 mm×1 mm were cut from different positions of bulk samples for superconducting properties measurement. A MPMS XL SQUID magnetometer was used to measure the magnetic moment as a function of temperature and the magnetic hysteresis loops of these specimens. J_c was calculated using the extended Bean model. For the measurements of trapped magnetic flux density, the bulk samples were cooled down to the liquid nitrogen temperature under the magnetic field of 1.0 T, which was applied parallelly to the c-axis. After 30 min from the removal of the external field, the automatic Hall probe scanning system with a F. W. Bell sensor, BHT-921 was employed to map the trapped magnetic flux density. The air gap distance between the top surface of the sample and the active area of the Hall sensor was 0.5 mm.

3 Results and discussion

3.1 Gd123 single grain processing

As shown in Fig. 2, Gd123 single grains are successfully processed by hot seeding, cold-seeding using Nd123/MgO thin film seed and cold-seeding using MgO-buffer pellet, respectively. Four growth folders

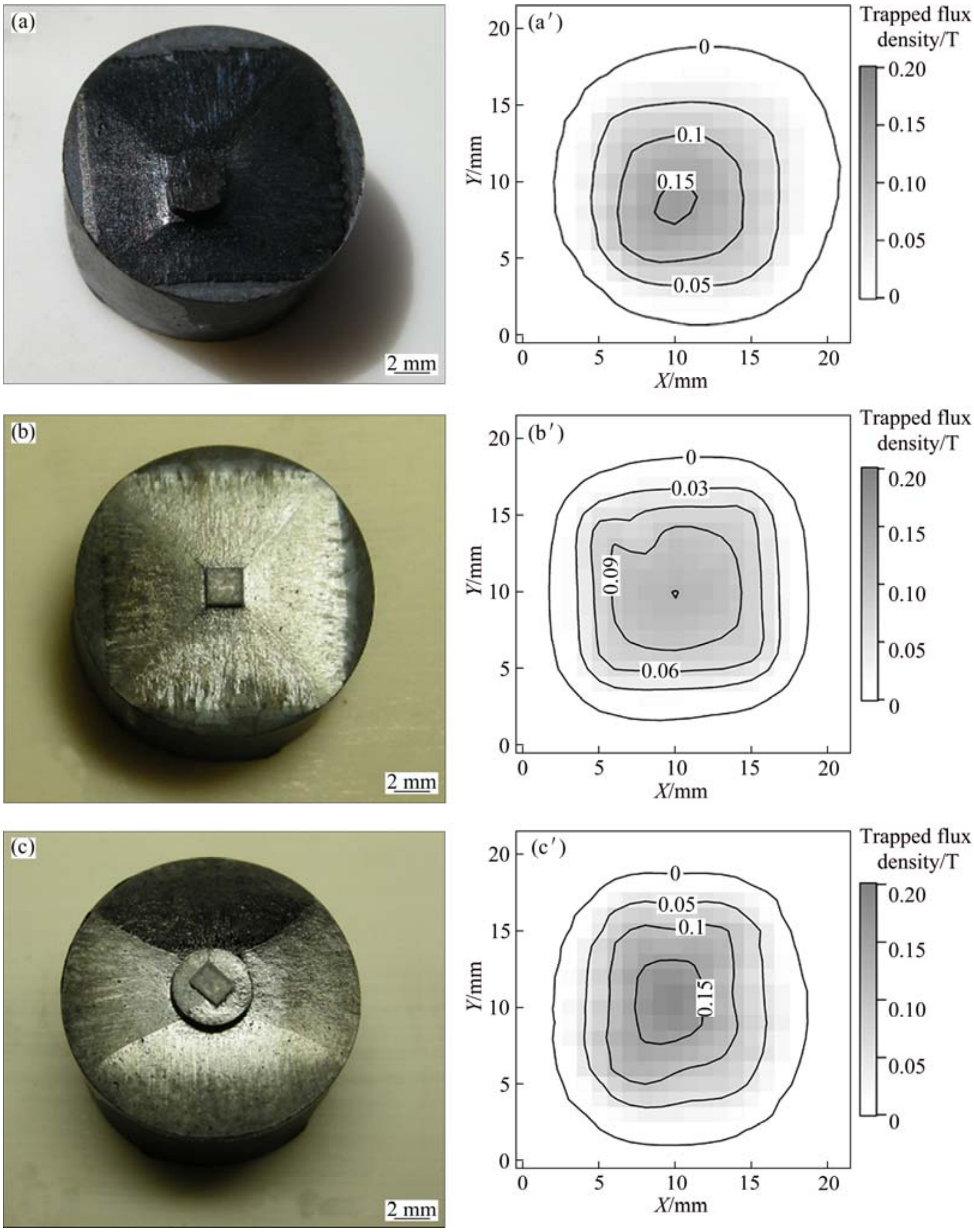

Fig. 2 Top view (a, b, c) and corresponding trapped flux density (a′, b′, c′) of Gd123 bulks processed by hot-seeding (a, a′), cold-seeding using Nd123/MgO thin film seed (b, b′) and cold-seeding using MgO-buffer pellet (c, c′)

originated from different growth sectors are clearly observed from the top view of each sample, indicating a quasi-single crystal growth. As one of the most important parameters for practical application, the trapped magnetic field was measured and the maximum flux densities ($B_{trapped}$) are 0.16, 0.15 and 0.19 T, respectively. This suggests that the cold seeding processing will not largely affect the superconducting performance of the Gd123 bulks, which is very important for the batch production.

However, the Nd123 thin films grown on MgO substrate still cost a lot. To realize the textured growth of Gd123 bulk from the MgO single crystal, one should overcome the problems of lattice mismatch and low reactivity between MgO and the matrix. In other words, one should realize the *c*-orientation growth from the MgO crystal, which largely depends on the growth rate of *a*/*b*- and *c*-axis. It is well known that growth rate is related to undercooling (ΔT), the difference between T_p and T_s, and the addition amount of RE211 particles. CARDWELL et al [15] reported that the geometry of NdBCO single grains, texture processed by MgO seeds, varied from rectangular (ΔT<10 °C) to rhombohedral (high ΔT) at different growth temperatures. The problem is to ensure the performance of the bulks. The composition of the matrix cannot be arbitrarily changed to meet the requirements of *c*-orientation growth. Therefore, we designed an additional step by inserting a buffer pellet. Figure 1(b) shows the results of the differential thermal analysis (DTA) from which the T_p is estimated. The large difference between T_p of precursor powder with Ag_2O and precursor powder without Ag_2O enables us to divide the growth into two steps. For the textured growth of the buffer pellet, we have carefully adjusted the undercooling from 10 to 25 °C, and the Gd211 content changes from 0% to 40% (mole fraction). Eventually, as shown in Fig. (2), a slow cooling between 1030 and 1025 °C with a rate of 0.5 °C/h and the Gd211 amount of 10% (mole fraction) are proved to be the suitable conditions for buffer pellet processing [16].

3.2 Doping effect of NiFe alloy particles

The cold seeding processing ensures the uniform growth conditions of all the Gd123 bulk samples, which is very important for a systematical research of the doping effect. The FeB particles doping may significantly enhance the J_c of Gd123 bulks in both self-field and intermediate field [13]. As a follow-up work, the NiFe alloy particle which is also named permalloy was chosen as candidate in the present work.

Figure 3 (a) shows the results of superconducting transition temperature (T_c) measurement of NiFe doped-samples with different doping amounts from 0.3% to 0.8% (mole fraction). T_c is suppressed with the increase of the doping amount, from 93.5 K for the no doping sample and from 90.5 K for the 0.8%-doped sample (mole fraction). But the superconducting transition width keeps sharp, about 1 K, even for the 0.8%- doped sample. Since both barium and oxide compounds of Fe and Ni are found in the quenched samples, it is suspected that the NiFe particles decomposed during the melt growth, although the NiFe alloy particles possess higher melting point than the T_{max}. Therefore, the suppression of T_c is supposed to be originated from the substitution of Fe and Ni ions for the Cu sites. It is well known that both Fe and Ni ions can easily substitute Cu ions at the Cu sites [17]. The solid solution limit of $GdBa_2(Cu_{1-x}Fe_x)O_\delta$ was reported to be 0.10, and the substitution usually occurs at CuO-chain [18]. For Ni ions, the solution limit was reported to be nearly 0.10 and the substitution occurs at CuO-plane [19].

As shown in Fig. 3 (b), J_c is dramatically enhanced under the external field larger than 0.5 T. The 0.4% NiFe doped sample shows the best performance, and a clear secondary peak can be observed at around 1.5 T. The volume pinning force F_p, suggesting the Lorentz force working on the magnetic flux ($J_c \times B$), versus the reduced irreversibility field H/H_{irr} is calculated based on the J—B curves. For all the NiFe doped samples, the h_0 is conformably equal to 0.42, which indicates that the contribution of NiFe doping is mainly δT_c pinning.

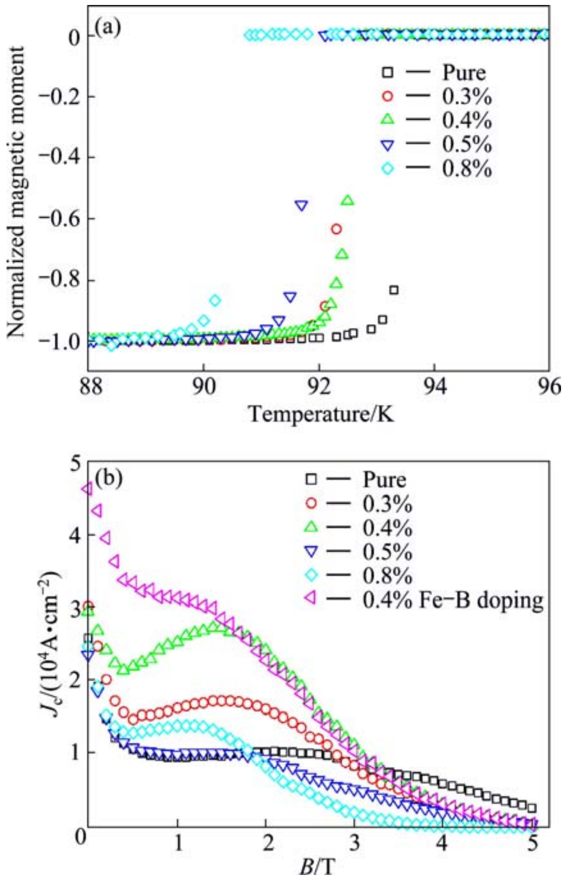

Fig. 3 T_c of bulk samples with different NiFe doping amounts (a) and J_c versus applied field B of bulk samples with different doping amounts (b)

For comparison, J_c—B curves of the Gd123 sample with optimal FeB doping amount is also exhibited in Fig. 3 (b). Refinement of the Gd211 particles and the size distribution have been observed by FeB doping [13]. However, for NiFe doping, no obvious refinements have been observed, which explains the low self-field J_c.

4 Conclusions

1) A novel cold-seeding method for TSMG bulk processing by combining MgO thin film and buffer pellet was reported. By adjusting the undercooling and Gd211 content of the buffer pellet, the nucleation problems originated from the lattice mismatch and low reactivity between Gd123 matrix and MgO seed are overcome. Since the melting point of MgO is sufficiently high, there is no T_{max} limitation for this method.

2) The doping effect of NiFe particles in the Gd123 bulks was studied. The 0.4% NiFe doped-sample shows the best flux pinning performance. A clearly secondary

peak in J_c—B curve is observed. The strong δT_c pinning is originated from the substitution of Fe and Ni for Cu sites.

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钆钡铜氧超导单晶块材的冷籽晶诱导生长及铁镍合金的掺杂作用

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摘 要：介绍了利用在 MgO 衬底上生长 $NdBa_2Cu_3O_y$(Nd123)薄膜从而诱导生长 $GdBa_2Cu_3O_y$(Gd123)超导单畴块材的工艺，并在此基础上研究了使用 MgO 缓冲层诱导生长钆钡铜氧单畴块材的工艺。通过调整热处理参数和缓冲层中 Gd_2BaCuO_5 (Gd211)相的含量，克服了 MgO 与 Gd123 母体的晶格失配和低反应率的问题。成功制备了 Gd123 单畴块材，研究了铁镍软磁合金粒子在超导块材中的掺杂作用。结果表明，0.4%(摩尔分数)为最优掺杂比例，超导临界电流在低场和中场下得到了很大的提升，对工业应用有重要意义。超导电流的提升主要源于铁、镍离子对铜位的替代，并提供了额外的磁通钉扎。

关键词：高温超导体；单晶生长；冷籽晶；化学掺杂；磁通钉扎；临界电流；二次峰效应

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